Oscillatory instability in super-diffusive reaction – diffusion systems: fractional amplitude and phase diffusion equations

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Nonlinear evolution of a reaction–super-diffusion system near a Hopf bifurcation is studied. Fractional analogues of complex Ginzburg-Landau equation and Kuramoto-Sivashinsky equation are derived, and some of their analytical and numerical solutions are studied.

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It has been recently realized that in many random physical processes the conceptions of Gaussian distribution and Fickian diffusion are invalid. Many such processes can be described by models of sub- or super-diffusion, where the displacement moments of the corresponding random walk grow slower or faster than for normal diffusion, respectively. A typical example of *super-diffusion* is the enhanced transport in fluids, predicted for flows with velocity correlation functions slowly decaying in space or time [1]. A specific type of super-diffusion, the Lévy flight, has been reported in observations of transport in two-dimensional rotating flows and in a freely decaying two-dimensional turbulent flow [2]. Other examples of super-diffusive transport include wave turbulence, non-local transport in plasma, transport in porous media, surfactant diffusion along polymer chains, cosmic rays propagation and motion of animals [3]. A widely used description of super-diffusive transport relies on the continuous time random walk model with a power law asymptotics of the particle jump length distribution, leading in the macroscopic limit to a diffusion equation with the Laplacian replaced by its *fractional power* [4].

An important problem is the influence of super-diffusion on processes with chemical reactions [5, 6]. Normal reaction– diffusion systems exhibit different types of instability [7]. Profound understanding of pattern formation and spatio-temporal chaos in these systems was achieved through generic equations valid near the instability threshold, such as complex Ginzburg-Landau [8] and Kuramoto-Sivashinsky equations [1]. The evolution of instabilities in reaction–diffusion systems can be accompanied by advection of components. For instance stirring, which changes the effective diffusion properties of species, is one of the means to control dynamical regimes generated by instabilities in reaction– diffusion systems [9]. Thus one can expect that in some cases flows can give rise to an enhanced diffusion of reagents. While studies of instabilities in systems with sub-diffusion have started (see [10] and references therein), super-diffusive reaction–diffusion systems are still unexplored, with the exception of the front propagation phenomenon, which is strongly influenced by fluctuations [6]. In this letter, weakly non-linear dynamics of a reaction–diffusion system characterized by Lévy flights near a long wave bifurcation point is investigated.

Consider a two-component reaction–diffusion system in the general case of distinct anomaly exponents:

$$\frac{\partial n_j}{\partial t} = d_j \mathfrak{D}^{\gamma_j}_{|x|} n_j + f_j(n_1, n_2), \quad j = \{1, 2\}, \tag{1}$$

where n_j, d_j and f_j are the species concentrations, diffusion coefficients and general kinetic functions, correspondingly. The fractional operator of order $1 < \gamma < 2$ is defined as [11]

$$\mathfrak{D}^{\gamma}_{|x|} n(x) = -\frac{\sec(\pi\gamma/2)}{2\Gamma(2-\gamma)} \frac{\partial^2}{\partial x^2} \int_{-\infty}^{\infty} \frac{n(\zeta)}{|x-\zeta|^{\gamma-1}} d\zeta. \tag{2}$$

The equivalent definition in Fourier space allows for a simple generalization of the operator to higher spatial dimensions: $\mathfrak{D}^{\gamma}_{|\mathbf{x}|} e^{i\,\mathbf{q}\cdot\mathbf{x}} = -|\mathbf{q}|^{\gamma} e^{i\,\mathbf{q}\cdot\mathbf{x}}$. Suppose that there exists a homogeneous steady state $\mathbf{n}_0$ satisfying $\mathbf{f}(\mathbf{n}_0) = \mathbf{0}$. A vanishing trace of the sensitivity matrix, $(\nabla \mathbf{f})_{jk} = \partial f_j / \partial n_k,\ j, k \in \{1, 2\}$, leads to Hopf bifurcation at the long wave limit $q = 0$. Take $\epsilon \ll 1$ and $0 < \mu \sim O(1)$ so that $\mathrm{tr}\, \nabla \mathbf{f}|_{\mathbf{n}_0} = \epsilon^2 \mu$ and invoke a multiple scales analysis with $\mathbf{n}(x, t) = \mathbf{N}(\xi, t_0, t_2, \ldots; \epsilon)$, $\xi = \delta(\epsilon) x$, $t_j = \epsilon^j t$, $j = 0, 2, \ldots$ and

$$\mathbf{N} \sim \mathbf{n}_0 + \sum_{j=1}^{\infty} \delta_j(\epsilon) \mathbf{N}_j(\xi, t_0, t_2, \ldots). \tag{3}$$

For normal diffusion ($\gamma = 2$) $\delta = \epsilon$, $\delta_j = \epsilon^j$ and a sequence of problems at successive orders δ_j is obtained. The solution at order δ_1 is of the form $\mathbf{N}_1 = A(\xi, t_2, \ldots) e^{i\omega_0 t_0} \mathbf{v}_1 + \mathrm{c.c.}$, where $\mathbf{v}_1$ is an eigenvector of the linearized problem and ω_0 is the Hopf bifurcation frequency. Neglecting the phenomena evolving on time scales longer than $\tau = t_2$, the equation for the amplitude A ensues as a solvability condition at order δ_3. For

an anomalous system the scaling property of the fractional operator, $\mathfrak{D}^{\gamma}_{|x|} y(x) = \delta^{\gamma} \mathfrak{D}^{\gamma}_{|\xi|} y(\xi/\delta)$, determines the scale of the slow spatial variable, δ. Namely, in a more common case with $\gamma_1 = \gamma_2 = \gamma$, the scale is $\delta = \epsilon^{2/\gamma}$ and $\delta_j = \epsilon^j$. The amplitude equation has the form of a *fractional complex Ginzburg-Landau* (FCGL) equation:

$$\frac{\partial A}{\partial \tau} = A + (1+\alpha i)\mathfrak{D}^{\gamma}_{|\xi|} A - (1+\beta i) A|A|^2 \qquad (4)$$

(in rescaled form). This equation was formerly derived in [12] in the problem of nonlinear oscillators' dynamics with long-range interactions. The parameters α and β coincide with those of a normal reaction – diffusion system but the Laplacian is replaced by the fractional operator. If $\gamma_1 \neq \gamma_2$, the super-diffusion term with the larger index is negligible in the long-wave region and $\delta_j = \epsilon^j$ for $j \leq 3$ only. Higher-order powers are fractional and depend on the ratio of the anomalous exponents. Then the appropriate scaling is $\delta = \epsilon^{2/\gamma}$ with $\gamma = \min\{\gamma_1, \gamma_2\}$, and the expressions for α and β are obtained by taking $d_2 = 0$ if $\gamma_1 < \gamma_2$ and $d_1 = 0$ if $\gamma_1 > \gamma_2$.

The integro-differential equation (4) retains the basic symmetries of a normal complex Ginzburg-Landau equation (with respect to time and space translations and the phase change $A \mapsto A\exp(i\vartheta)$). It is interesting that its solutions in the form $A(\xi,\tau) = B(\xi)\, e^{i(q\xi - \omega\tau)}$, with $q, \omega \in \mathbb{R}$, have a symmetry similar to that found by Hagan [13]. If a solution of this type is known for a pair (α, β), the solution for a new pair (α', β') located on one of the curves $(\alpha-\beta)/(1+\alpha\beta) = \text{const}$ can be found by the transformation $B = aB'$, $\xi = b\xi'$, where

$$a^2 b^{\gamma} = \frac{1+\alpha'\beta'}{1+\alpha\beta}\frac{1+\alpha^2}{1+\alpha'^2}, \qquad (5a)$$

$$b^{\gamma} = \frac{1+\alpha^2}{1+\alpha\alpha' + (\alpha-\alpha')\omega}, \qquad (5b)$$

and the new wavenumber and frequency are $q' = bq$, $\omega' = \alpha' - b^{\gamma}(1+\alpha'^2)(\alpha-\omega)/(1+\alpha^2)$.

In the special case $\alpha = \beta$ eq. (4), after the phase shift $A \mapsto A\exp(-i\beta\tau)$, eq.(4), like a normal complex Ginzburg-Landau equation [14], can be written in a variational form,

$$\frac{\partial A}{\partial \tau} = -(1+i\beta)\frac{\delta \Upsilon}{\delta A^*}, \qquad (6)$$

where $\Upsilon = \int_{-\infty}^{\infty} U(\xi,\tau) d\xi$, and

$$U = -|A|^2 + \frac{|A|^4}{2} - \frac{\sec(\pi\gamma/2)}{2\Gamma(2-\gamma)}\left\{\frac{\partial A^*}{\partial \xi}\frac{\partial}{\partial \xi}\int_{-\infty}^{\infty}\frac{A(\zeta)d\zeta}{|\xi-\zeta|^{\gamma-1}}\right.$$

$$\left. + \frac{1-\gamma}{2} A \int_{-\infty}^{\infty}\frac{\partial A^*}{\partial \zeta}\frac{\text{sign}(\xi-\zeta)}{|\xi-\zeta|^{\gamma}} d\zeta + \text{c.c.}\right\} + c. \qquad (7)$$

The constant c is chosen so that Υ converges. Then $\partial\Upsilon/\partial\tau = -2(1+\beta^2)^{-1}\int_{-\infty}^{\infty} |\partial A/\partial t|^2\, d\xi < 0$, and the system relaxes to a certain stable "stationary" solutions (the original variable A oscillates with the frequency β).

Now consider the traveling wave solutions of (4),

$$A_q = \sqrt{1-|q|^{\gamma}} e^{i(q\xi-\omega\tau)}, \;\; \omega = \beta - (\beta-\alpha)|q|^{\gamma}. \qquad (8)$$

A small perturbation $a(\xi,\tau)$ about A_q comprises longitudinal and transverse waves of the form

$$a = A_{q+k}(\tau) e^{i(q+k_\xi)\xi + ik_\eta\eta} + A_{q-k}(\tau) e^{i(q-k_\xi)\xi - ik_\eta\eta}, \qquad (9)$$

with k_ξ, k_η being the respective wave numbers. The solution (8) is neutrally stable with respect to disturbances $k_\xi = k_\eta = 0$. Further insight into long perturbations reveals that for $O(k_\xi/q) \sim O(k_\eta/q) \sim o(1)$ to leading order the growth rate of $A_{q\pm k} \sim \exp(\lambda\tau)$ satisfies

$$\Re\lambda \sim \frac{\gamma}{2}|q|^{\gamma}\left[-(1+\alpha\beta)\left((\gamma-1)\frac{k_\xi^2}{q^2} + \frac{k_\eta^2}{q^2}\right) + \right.$$

$$\left. \gamma(1+\beta^2)\frac{|q|^{\gamma}}{1-|q|^{\gamma}}\frac{k_\xi^2}{q^2}\right]. \qquad (10)$$

Therefore all solutions (8) are unstable if $1+\alpha\beta < 0$, i.e. the Benjamin-Feir criterion for a normal CGLE is recovered. However, if $1+\alpha\beta > 0$, a γ-dependent set of unstable wave vectors exists, generalizing the Eckhaus instability criterion:

$$|q_m| < |q| < 1, \;\; |q_m|^{-\gamma} = 1 + \frac{\gamma}{\gamma-1}\frac{1+\beta^2}{1+\alpha\beta}. \qquad (11)$$

No new instability criteria emerge in the opposite limit $q \ll k_\xi, k_\eta \ll 1$. In particular, the spatially-homogeneous oscillation $A_0 = \exp(-i\beta\tau)$ is unstable in the same region $1+\alpha\beta < 0$ with respect to disturbances whose wave numbers k satisfy

$$0 < |k|^{\gamma} < -\frac{2(1+\alpha\beta)}{(1+\alpha^2)}, \qquad 1+\alpha\beta < 0. \qquad (12)$$

The evolution of perturbations near the domain boundary is expected to be described by an analogue of the Kuramoto-Sivashinsky equation [1]. Define $1+\alpha\beta = -\epsilon$, $\;0 < \epsilon \ll 1$, rewrite (4) with $\chi = \epsilon^{1/\gamma}\xi$ and $\tau_2 = \epsilon^2\tau$, take $A = \exp(-i\beta\tau_2/\epsilon^2)\, r(\chi,\tau_2)\, \exp[i\varphi(\chi,\tau_2)]$, where $r = 1 + \sum_{j=1}^{\infty}\epsilon^j\, r_j(\chi,\tau_2)$, $\varphi = \sum_{j=1}^{\infty}\epsilon^j\,\varphi_j(\chi,\tau_2)$, and expand $\exp(\pm i\varphi)$ to obtain the phase diffusion equation at order $O(\epsilon^3)$ that, after rescaling, has the following form (notations for the rescaled space and time variables are the same):

$$\frac{\partial \phi}{\partial \tau} = -\mathfrak{D}^{\gamma}_{|\chi|}\phi - (\mathfrak{D}^{\gamma}_{|\chi|})^2\phi + \frac{1}{2}\mathfrak{D}^{\gamma}_{|\chi|}\phi^2 - \phi\mathfrak{D}^{\gamma}_{|\chi|}\phi. \qquad (13)$$

The operator $(\mathfrak{D}^{\gamma}_{|\chi|})^2$ is defined in Fourier space by $(\mathfrak{D}^{\gamma}_{|\chi|})^2 e^{iq\chi} = |q|^{2\gamma} e^{iq\chi}$ and cannot be simply related to

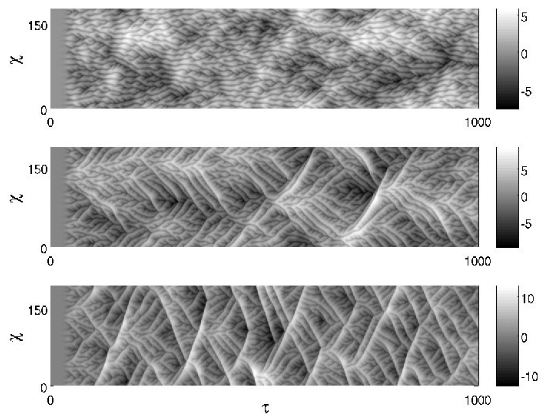

Figure 1: Spatio-temporal dynamics of solutions of eq.(13) for $\gamma = 2.0$ (upper), $\gamma = 1.7$ (middle), and $\gamma = 1.6$ (lower).

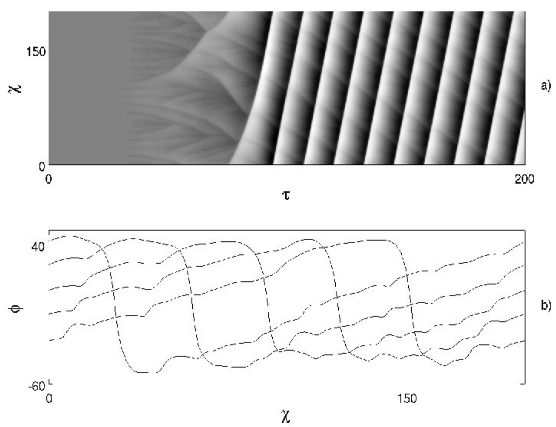

Figure 2: (a) Spatio-temporal dynamics of solutions of eq.(13) for $\gamma = 1.5$. (b) Solutions of (13) at successive moments of time.

the operator $\mathfrak{D}^{2\gamma}_{|\chi|}$ as the order 2γ exceeds the definition range in (2). Eq.(13) is the *fractional Kuramoto-Sivashinsky equation.*

Fig.1 shows spatio-temporal dynamics of the numerical solutions of eq.(13) obtained by means of a pseudospectral code, using periodic boundary conditions and starting from small-amplitude random data. The upper figure shows the dynamics for $\gamma = 2$ corresponding to a normal KS equation: this is a well-known spatio-temporal chaos exhibiting merging and splitting of "cellular" structures [1]. The middle figure corresponds to $\gamma = 1.7$. One can see that along with the chaotic dynamics of "cells" large-amplitude traveling "shocks" develop that emit cells still displaying chaotic dynamics. With further decrease of γ, the shocks appear more frequently, propagate faster and their amplitude grows, see the lower figure corresponding to $\gamma = 1.6$. When γ decreases below a certain threshold that depends on the domain length, a single traveling shock is formed in the whole domain. An example of such a shock is shown in Fig.2. Here the shock is trav-

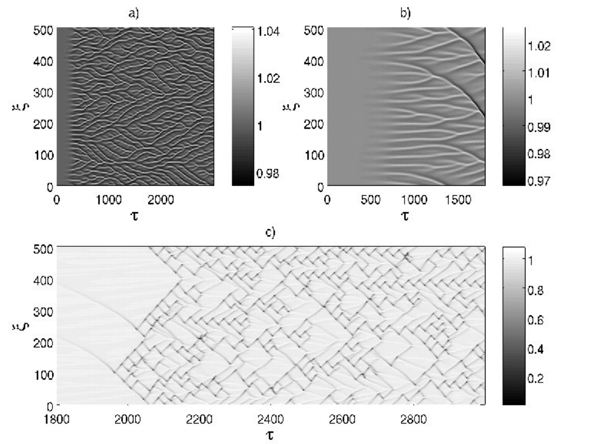

Figure 3: Spatio-temporal dynamics of solutions of eq.(4) for (a) $\alpha = -1,\ \beta = 1.33,\ \gamma = 2.0$; (b) $\alpha = -1,\ \beta = 1.2,\ \gamma = 1.6$; (c) continuation of (b) for the same parameter values.

eling with a constant speed (Fig.2a) while its "wings" exhibit spatio-temporally chaotic modulations, (Fig.2b). Decreasing γ results in the increase of the shock amplitude and after certain critical γ the shock starts accelerating with its amplitude growing exponentially. The shock amplitude grows with the size of the computational domain. An asymptotic analysis carried out for large-amplitude solutions of (13) shows that the solution is of the form $\phi = a(\tau) f(\xi - \zeta(\tau))$, where f is an odd periodic function and $a(\tau)$ grows exponentially (despite the problem non-linearity) with a certain dependence on the domain size and the velocity $d\xi(\tau)/d\tau$ proportional to $a(\tau)$. The numerical simulations confirm the asymptotic analysis.

Next, numerical simulations of the FCGL equation (4) in 1D have been performed for the phase turbulence regime. Fig.3a shows spatio-temporal dynamics typical of the normal CGL equation, starting from the Benjamin-Feir-unstable, spatially-homogeneous oscillations: it is well described by the normal KS equation (see Fig.1a). Fig.3b shows the similar dynamics of eq.(4) for $\gamma = 1.6$. One can see that, after some period of phase turbulence, accelerating shocks form that trigger the transition to defect turbulence shown in Fig.3c. The formation of the accelerating shocks seen in Figs.3b,c is consistent with the formation of shocks in the FKS equation discussed above.

Fig.4 shows the spatio-temporal dynamics of numerical solutions of eq.(4) corresponding to the defect turbulence regime emerging from the Benjamin-Feir-unstable wave (8) with $q = 0.5$ for $\gamma = 2.0$ and $\gamma = 1.1$ (Figs.4 (a) and (b), respectively). One can see that in the anomalous case the defect turbulence has a stronger phase-turbulence component and does not consist of propagating holes.

Finally, numerical simulations of FCGL eq.(4) in 2D

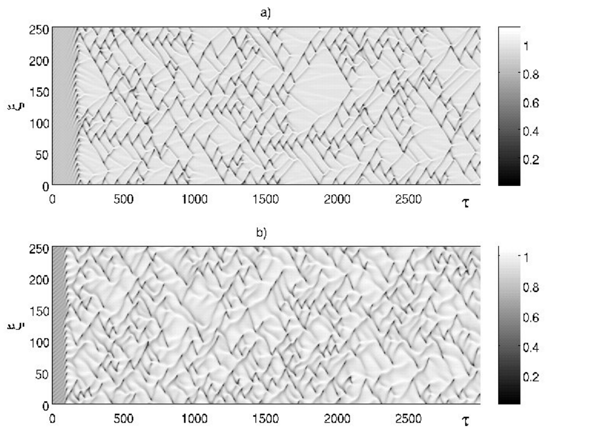

Figure 4: Spatio-temporal dynamics of $|A|$ – solution of eq.(4) for $\alpha = 1$, $\beta = -1.3$ and (a) $\gamma = 2.0$; (b) $\gamma = 1.1$.

have been performed for the parameter values corresponding to the formation of spiral waves in the normal CGL equation. Periodic boundary conditions and small-amplitude random initial data were used. The results are shown in Fig.5. One can see that for γ close to 2 (see Figs.5a,d) the formation of a spiral wave is still observed. With the decrease of γ the spiral-wave regime is replaced by a defect chaos, however, remnants of the spiral waves still can be seen (Figs.5b,e), with each "spiral" occupying a small domain with the domain walls partially melted. Further decrease of γ results in the decrease of the number of defects, the domain walls are almost completely melted (Figs.5c,f), and the local wavenumber created by each defect decreases. A "phase diagram" of the new dynamical states described above in the parameter space will be presented elsewhere.

In conclusion, we have derived fractional Ginzburg-Landau and Kuramoto-Sivashinsky equations that describe weakly non-linear dynamics of a super-diffusive reaction–diffusion system, characterized by Lévy flights,

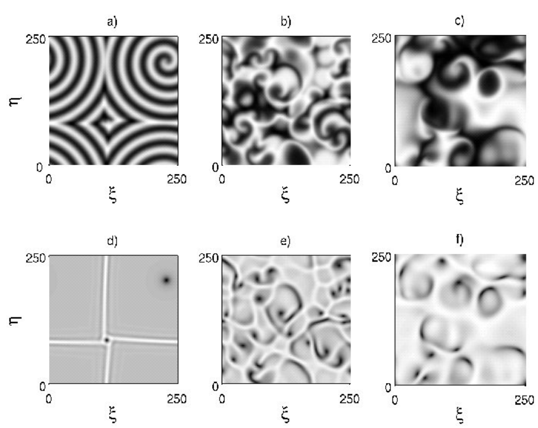

Figure 5: Snapshots of solutions of eq.(4) for $\alpha = 1.5$, $\beta = -0.6$ and $\gamma = 1.9$ (a),(d); $\gamma = 1.8$ (b),(e); $\gamma = 1.05$ (c),(f); upper figures – Re(A), lower ones - $|A|$.

and studied some of their solutions analytically and numerically. We have shown that super-diffusion can lead to a transition from phase- to defect turbulence and to destruction of spiral waves. We note that investigating the effects of fluctuations on non-linear dynamics of instabilities in a super-diffusive reaction–diffusion system would be of interest since, as shown in [6], the fluctuations can have a profound influence on the non-linear behavior in such systems. However, this topic is beyond the scope of the present paper.

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